May 1990

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We have measured the magnetization and magneto-optic Kerr rotation of sputtered thin films of Mn-Al-Cu ternary alloys over a wide composition range and for substrate temperatures between 25 and 450 °C. Magnetization and x-ray diffraction measurements suggest that our films are a mixture of the ferromagnetic kappa (κ) phase of MnAl and a highly paramagnetic amorphous phase. Room-temperature magneto-optic measurements on these films show a saturation polar Kerr rotation of up to 0.12° with little wavelength dependence.

INTRODUCTION

Since several Mn-based materials, most notably MnBi (Ref. 1) and PtMnSb (Ref. 2), have large Kerr rotations, we have been investigating the magneto-optical properties of other Mn-based alloys. In particular, our earlier work on evaporated Mn-Bi-Al films showed anomalous hysteresis loops that could be treated as arising from two different (and magnetically independent) magnetic phases.[3] One of these phases was the familiar MnBi,[4] with a Kerr rotation in excess of 1°, while the other appeared to be the metastable MnAl τ phase.[5–8] Our analysis of the Kerr rotation hysteresis loops on these materials led us to conclude that the MnAl τ phase has a Kerr rotation similar in magnitude to that of MnBi, so that the τ phase suggested itself as an interesting system for further study. Thin films of the τ phase have been shown to be stabilized by the addition of Cu.[8] Similarly, the ferromagnetic MnAl-based kappa (κ) phase has been prepared in both bulk and thin-film forms, with this phase also being stabilized by the addition of Cu.[8,9] In view of these results, we have prepared thin films of the Mn-Al-Cu system by magnetron sputtering over a wide composition range, and we have investigated their magnetic and magneto-optic properties.

Tsuboya has shown that the bulk κ phase can be stabilized by incorporating Cu into the melt.[9] The structure of the κ phase is CsCl-type bcc, with formula (Mn,Cu)Al. More recently, Morisako, Matsumoto, and Naoe have produced $Mn_{60-x}Al_{40}Cu_x$ sputtered films with both the κ- and τ-phase structures.[8] For $x < 10$, the films were composed of the τ phase, while for $10 < x < 30$, the κ phase was stable. Keeping the Al concentration at 40 at. %, they found a maximum room-temperature saturation magnetization of 300 emu/cm^3 for a Mn concentration of 38 at. %.

In this paper, we report results of our studies of the magnetic and magneto-optic properties of thin films in the Mn-Al-Cu ternary alloy system. The major magnetic phase produced in this work is the cubic (bcc) κ phase.

EXPERIMENT

The thin films were deposited by magnetron sputtering in a chamber of base pressure 2×10^{-7} Torr and with an Ar gas pressure of 5 mTorr. The films were deposited on Pyrex substrates at substrate temperature T_s ranging from 20 to 450 °C. The compositions of the films were varied by changing the number of Mn and Cu chips on the Al sputtering target. The film compositions were determined by x-ray fluorescence, and their structures were determined by x-ray diffraction. The magnetizations were measured using a superconducting quantum interference device (SQUID) magnetometer, while the polar Kerr rotation measurements were made using a homemade magneto-optic apparatus.[3]

RESULTS AND DISCUSSION

We produced films with Mn concentrations ranging from 10 to 55 at. % and Cu concentrations up to 20 at . %. X-ray diffraction and Kerr rotation measurements revealed no evidence for the metastable τ phase in any of these films, in contrast to the work of Morisako, Matsumoto, and Naoe.[8] They found that the τ phase was stabilized with Mn concentrations near 50 at. % and Cu concentrations below 10 at. %. Our films with compositions in this range were always found to be amorphous and nonmagnetic. X-ray diffraction scans for $Mn_{27}Al_{61}Cu_{12}$ are shown in Fig. 1 for several substrate temperatures. For $T_s < 50$ °C, the film is amorphous and nonmagnetic. For $50 < T_s < 130$ °C, two phases, the nonmagnetic ϵ phase and the ferromagnetic κ phase, coexist. The (002) diffraction peak of the ϵ phase is coincident with the (111) peak of the τ phase, but the phase present in our films is nonmagnetic, indicating that it is indeed the ϵ phase. As T_s approaches 130 °C, the ϵ phase (002) peak gradually disappears, so that for $130 < T_s < 250$ °C, only the κ phase is present. For substrate temperatures above 250 °C, the films are again amorphous.

Figure 2 shows the magnetization obtained at 30 kOe for $Mn_{27}Al_{61}Cu_{12}$ ($T_s = 200$ °C) for temperatures between 5 and 400 K. The diamagnetic contribution from the Pyrex substrate has been subtracted from the raw measurements, assuming that the Pyrex contribution was independent of temperature. The x-ray diffraction measurements on this sample showed only the κ-phase (110) peak. The saturation magnetization of 32 emu/g at room temperature is about one half the value found by Morisako, Matsumoto, and Naoe in their thin-film κ-phase samples.[8] Tsuboya found that both the saturation magnetization and the Curie temperature of the κ phase were quite composition dependent, with Curie temperatures up to 470 K. As can be seen in Fig. 2, the Curie

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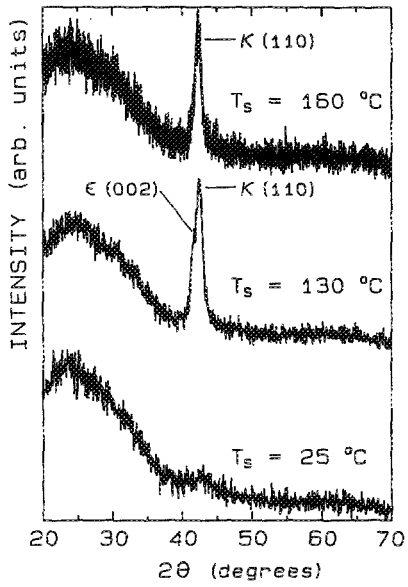

FIG. 1. X-ray diffraction scans for $Mn_{27}Al_{61}Cu_{12}$ films deposited at different substrate temperatures T_s.

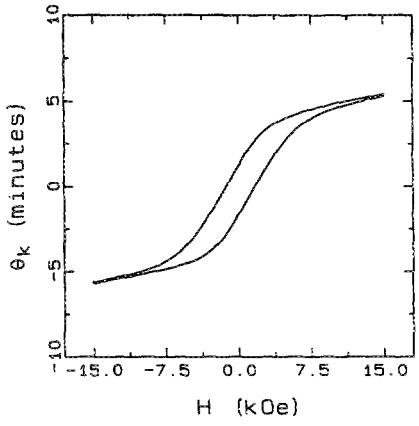

FIG. 3. Polar Kerr rotation hysteresis loop for $Mn_{27}Al_{61}Cu_{12}$ ($T_s = 200$ °C). The magnetic field was applied perpendicular to the film, and the measurements were taken at 300 K. The light source was a He-Ne laser operating at 633 nm.

temperature for our sample lies somewhat above 400 K. The rise in magnetization at low temperature can be fitted quite well by a Curie law behavior, suggesting that a highly paramagnetic impurity phase is also present in the film. The lack of x-ray structure suggests that this phase is amorphous. Low-temperature SQUID measurements on amorphous films of similar composition that were nonmagnetic at room temperature also showed this paramagnetic behavior. Morisako, Matsumoto, and Naoe found that the moment per Mn atom in the bulk κ phase was $0.87\mu_B$,[8] or considerably less than the moment of an isolated Mn ion, which is typically $4\mu_B$ to $6\mu_B$. In view of this, we suggest that roughly one-half of our sample is κ phase, with the other half being an amorphous phase containing relatively few Mn ions of relatively large magnetic moment.

Figure 3 shows the polar Kerr rotation hysteresis loop (at 633 nm) of $Mn_{27}Al_{61}Cu_{12}$ deposited at a substrate temperature of 200 °C. The magnetic field was applied perpendicular to the film, and the sample was at room temperature. Only the κ-phase (110) diffraction line is observed in this film. The saturation value of the Kerr rotation is about 6 min and the coercivity is 1.7 kOe, with the latter value being in reasonable agreement with the coercivity obtained by Morisako, Matsumoto, and Naoe.[8]

Figure 4 shows the saturation Kerr rotation (at 633 nm) and coercivity of $Mn_{27}Al_{61}Cu_{12}$ as a function of substrate temperature. Both quantities increase in the same manner, reaching a peak near 200 °C, and finally dropping to zero near 250 °C. The lower-temperature behavior is likely due to the gradual disappearance of the ϵ phase as the temperature is raised to 130 °C. The behavior above 200 °C is most likely due to the disappearance of the κ phase in favor of an amorphous phase. The maximum Kerr rotation, obtained for a substrate temperature of 200 °C, is about 6 min. This value should be considered only an approximate value for the κ phase, because of the presence of a second (amorphous) phase, which is paramagnetic at the measurement temperature. One might speculate that if our films were pure κ phase, the Kerr rotation would be about twice as large.

Figure 5 shows the wavelength dependence of the satu-

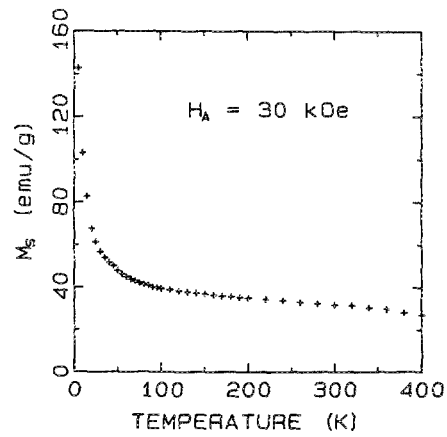

FIG. 2. Magnetization of $Mn_{27}Al_{61}Cu_{12}$ ($T_s = 200$ °C) with a 30-kOe applied field as a function of temperature.

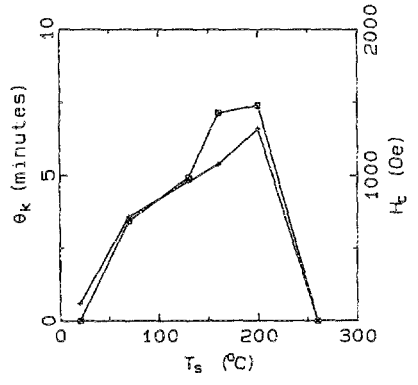

FIG. 4. Saturation Kerr rotation (□) and coercivity (+) in $Mn_{27}Al_{61}Cu_{12}$ as a function of substrate temperature. The measurements were made at 300 K.

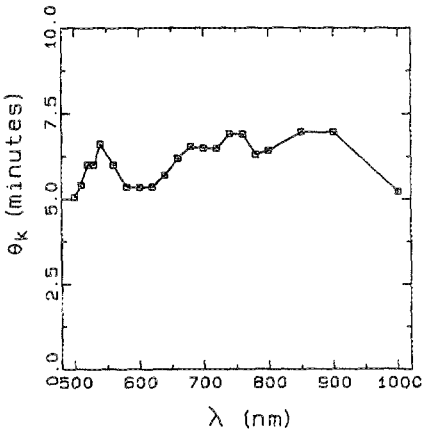

FIG. 5. Saturation Kerr rotation $Mn_{27}Al_{61}Cu_{12}$ as a function of incident light wavelength. The measurements were made at 300 K.

ration Kerr rotation for $Mn_{27}Al_{61}Cu_{12}$ ($T_s = 200\,^{\circ}C$). The magnitude of the Kerr rotation is only weakly wavelength dependent, with a value near 6 min.

In summary, we have studied sputtered thin films of the system Mn-Al-Cu over a wide composition range and for a variety of substrate temperatures. The only magnetic phase observed is the bcc structure κ phase, but our films appear to contain a highly paramagnetic amorphous phase as well. The films show a relatively small Kerr rotation, which is not surprising in view of their small saturation magnetization and in view of the lack of any elements that would lead to a large spin-orbit coupling.[10] The reasons for the differences in structure between our films and those of Morisako, Matsumoto, and Naoe[8] are not clear, but some possibilities include different substrate temperatures or the presence of unknown impurities such as oxygen in one of the preparation chambers. Further work on isolating the τ phase in bulk and thin-film samples is underway.

ACKNOWLEDGMENTS

We wish to thank S. H. Liou for help with the magnetization measurements and S. S. Jaswal for helpful discussions. This research was supported by the NSF under Grants No. DMR-8605367 and No. INT-8715441. Additional support from Vice Chancellor Yost and the NU Foundation is also greatly appreciated.

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